

REVIEW

Polypropylene-based nanocomposites for HVDC cable insulation

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Abstract

Cross-linked polyethylene (XLPE) is commonly used as an insulation material in power cables. Due to the recent advancements in the field of high voltage power transmission and distribution, there is a need for novel cable insulation materials that have high performance, recyclability and high working temperature as alternatives for the conventional XPLE-based insulation materials. Polypropylene (PP) shows excellent properties and has drawn considerable attention as a potential high voltage direct current (HVDC) insulation material. Therefore, the development of PP-based HVDC cable insulation with improved electrical, thermal and mechanical properties is important in discovering a potentially recyclable cable insulation material. Due to the remarkable development in the field of nanodielectrics, nanotechnology can be a promising solution for enhancing the overall dielectric properties of PP-based insulation materials. This review presents the important aspects of PP-based nanocomposites for HVDC cable insulation with a special focus on understanding the effects of various parameters of nanofillers on the dielectric properties of PP-based HVDC cable insulation. Based on the gathered information, future perspectives for improving the dielectric properties of PP-based nanocomposites for HVDC cable are provided.

1 | INTRODUCTION

High voltage direct current (HVDC) cable transmission has drawn increased attention due to the great development in high voltage power transmission technologies in recent years. Currently, polymer-based materials are largely used in various high voltage apparatuses such as power transformers, insulators, capacitors, reactors, surge arresters, current and voltage sensors, power cables and terminations. Cross-linked polyethylene (XLPE), for example, has been widely adopted as an insulating material for high voltage power cables owing to its good properties such as higher working temperature, low dielectric loss, enhanced tree resistance and better chemical and oil resistance [1]. Due to these enhanced properties, XLPE has been the choice for HVDC cable insulation materials over last decades. However, XLPE suffers some serious drawbacks as an insulating material for HVDC cables such as undesirable space charge accumulation, long processing time, high production cost and non-recyclability. Therefore, in view of increasing demand for energy

and sustainability, alternatives for traditional XLPE-based materials have been looked into.

With the rapid development of HVDC cable insulation materials, the need for higher operating voltage and large power capacity at reduced cable size, weight and volume has arisen [2]. In recent years, due to their superior thermal, electrical and mechanical properties, polypropylene (PP)-based insulating materials have gained significant interest from both academia and industry for replacing XLPE-based insulating materials in power cable insulation [3–6]. The melting point of PP can be as high as 170°C, which is 40–50% higher than that of polyethylene (PE). Due to its high melting temperature, the long-term working temperature of PP can easily be extended to 100–120°C. Moreover, due to its hydrophobic nature, humidity has less effect on its insulation properties. Regarding its electrical properties, PP possesses high breakdown strength, low dielectric constant, low dielectric permittivity and high DC volume resistivity [7]. Besides that, PP has high mechanical strength, which means that it can be manufactured without any

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crosslinking treatment. So, it can be recycled and reused at ease at the end of its service life compared to conventional XLPE [8]. These properties make PP-based material favourable for HVDC cable insulation.

Meanwhile, nanodielectrics have recently emerged as a promising solution in improving the overall properties in polymer-based insulating materials. Nanodielectrics, sometimes referred as nanocomposites, are a class of hybrid organic–inorganic materials, in which a small amount of nanofillers with at least one dimension in the range of 1–100 nm are homogeneously dispersed in polymers [9]. The concept of nanometric dielectric in electrical insulation was brought about by Lewis in 1994 by exploring the role of nanometric size particle inclusion in polymers that would lead to the enhancement in material properties [10]. It is believed that the improved properties of nanocomposites are attributed to the huge interfacial region formed between the nanofillers (high surface area) and the polymers [11]. In order to explain the effect of the interfacial region, several theoretical models of nanodielectrics have been proposed and compared.

It has been shown that addition of different nanofillers into PP resulted in enhanced dielectric properties. Notably, space charge, dielectric breakdown strength and DC conductivity are considered as important electrical parameters in high voltage insulation systems. Therefore, the development of PP-based HVDC cable insulation with effective space charge suppression, high breakdown strength and lowered DC conductivity is important in realizing its potential as cable insulation materials. Till date, a small amount of different kinds of nanofillers like zero-dimensional (0D), one-dimensional (1D) and two-dimensional (2D) nanofillers are incorporated into PP-based polymer to improve its overall properties. For example, the inclusion of nanofillers in PP resulted in improved thermal conductivity and stability [12–14], effective space charge suppression [15–17], enhanced partial discharge resistance [18, 19], high volume resistivity [20–22] and suppressed electrical treeing [23]. These enhanced properties are related to the presence of deep traps at the polymer/nanofiller interface that helps in the capturing and scattering of free charge carriers injected from high voltage electrodes [24, 25].

Thus far, numerous works have been published on enhancing the overall properties of PP-based nanocomposites for HVDC cable insulation. As far as the authors are aware, there is limited study that specifically compiles the work related to the effects of different parameters of nanofillers on the dielectric properties of PP-based nanocomposites. In this review, we outline the limitations of conventional XLPE insulation materials, followed by brief discussion on the structure, advantages, and properties of PP-based materials. This is followed by discussion on various kinds of nanofillers used for improving the dielectric properties of PP materials for HVDC insulation and discussion of various surface modification techniques used in modifying the surface of nanofillers. Afterwards, different theoretical nanodielectrics models are discussed and compared. Subsequently, different fabrication methods for polymer nanocomposites are discussed. Finally,

various parameters of nanofillers (such as nanofiller loading, types, surface chemistry and morphology) and temperature that influence the dielectric breakdown strength of PP nanocomposites are reviewed. Lastly, some future perspectives for tailoring the properties of PP nanocomposites for HVDC cable insulation materials are provided.

2 | LIMITATION OF CONVENTIONAL XLPE

Over the past 60 years, XLPE has been widely adopted as an insulating material for high-voltage power cables owing to its good mechanical and electrical properties. XLPE is obtained by crosslinking of polyethylene (PE) using crosslinking agents. Dicumyl peroxide is a typical crosslinking agent that is used to crosslink PE that results in three dimensional crosslinked polymer network with enhanced thermochemical properties than the base material (PE). The crosslinking process of PE resulted in improved stability, with the operation temperature of XLPE improved to 90°C as proposed to 70°C for PE [26]. However, in view of an increasing demand for energy, traditional XLPE-based insulation materials face problems as these materials are rated to operation temperatures of about 90°C. A further increase in its energy capacity will increase its working temperature beyond 90°C, which can result in fast ageing and final breakdown of the material, thus limiting its use for high operating voltages. In addition, the involvement of crosslinking agents, typically dicumyl peroxide, yields unwanted by-products such as methane, phenol, acetophene, 2-methylstyrene and cumyl alcohol. The presence of these by-products in XLPE cables can result in space charge accumulation under DC electric field [27, 28]. The accumulated space charge in XLPE can lead to the distortion of local electric field within the insulation material. As a result, electrical degradation can occur which may lead to insulation failure [29]. Although these by-products can be removed from XLPE cables by degassing during production process, the long processing time and high temperature for degassing increase the production time and cost [30]. Furthermore, crosslinking makes XLPE a thermosetting material that is difficult to be recycled by the end of its service life and thus cannot be reused. Taking into consideration the aforementioned limitations of XLPE, it is therefore highly desirable to develop an environmental friendly HVDC cable insulation material which has superior properties over XLPE.

3 | POLYPROPYLENE AS A POTENTIAL HVDC CABLE INSULATION MATERIAL

PP belongs to the group of polyolefins which can be obtained through the process of polymerization of propylene monomers. PP can be found in three stereo specific configurations based on the attachment of methyl group (–CH₃) on the polymer backbone [31] specifically, isotactic PP (iPP) having methyl groups on one side of polymer backbone, syndiotactic PP (sPP) having methyl groups in alternative order on polymer

backbone and atactic PP (aPP) having irregular attachment of methyl groups on the polymer backbone. These are illustrated in Figure 1. iPP and sPP are semi crystalline while aPP is amorphous polymer which has no practical usage. Based on chemical bonding, PP can be divided into homopolymer PP (HPP) containing only propylene monomer, which is also referred as iPP, random copolymer PP (rPP) containing ethylene in the PP chains at about 1–8% and impact copolymer containing a mixture of HPP and rPP with an overall content of 45–65% of ethylene content.

Regarding the basic properties requirements for HVDC cable insulation materials, the intrinsic high melting temperature of PP allows it to carry high voltages and withstand higher working temperature thus avoiding the use of crosslinking agents. Second, PP does not have impurities problem like XLPE and this favours dielectric properties of PP [2]. Thus, with respect to XLPE, PP shows the advantages of enhancing thermal and electrical properties, no by-products formation, no degassing treatments requirement and is recyclable. Yoshino et al. [31] did a comparative study on the AC breakdown strength of sPP, iPP and XLPE where they found that the AC breakdown strength of sPP was much higher than iPP and XLPE. Although sPP has shown better properties for HVDC cable insulation, its high manufacturing cost limits its wide range application in HVDC cable insulation.

Recently, iPP has drawn much attention as a potential material for HVDC cable insulation due to its higher breakdown strength and low manufacturing cost. However, its high stiffness and brittleness at low temperature hinder its use in cable insulation. Nevertheless, different strategies have been adopted to overcome its limitations. Copolymerization is a useful technique to tailor the mechanical, electrical and thermal properties of PP. Propylene monomers can be processed with other polyolefins such as ethylene or butene to produce PP copolymers. Different studies have investigated the electrical, thermal and mechanical properties of various PP copolymers. For example, Huang et al. [32] evaluated the potential of PP materials for HVDC cable insulation. Six different PP polymers (two iPP, two propylene-ethylene block copolymer and two propylene-ethylene random copolymer (rPP)) were considered. Among the mentioned polymers, rPP showed best properties in terms of mechanical flexibility, high melting point, reduced space charge and high breakdown strength. Their results pointed out that rPP can be a potential material for HVDC cable insulation. Hosier et al. [33, 34] investigated PP ethylene copolymers and their blends. Their results showed that the mechanical properties of the materials increased but at the expense of reduced breakdown strength.

Blending PP with other polymers is also used to alter the properties of virgin PP. Various polymers like ethylene propylene rubber, ethylene propylene diene monomer (EPDM) and polyolefin elastomers (POEs) have been blended with PP for its property improvements. Hosier et al. [4] investigated the thermal, mechanical and electrical properties of different iPP/sPP and iPP/propylene-ethylene copolymer (PEC) blends. Their investigation indicated that among different blends, iPP/PEC blend containing 40 wt% ethylene showed optimized

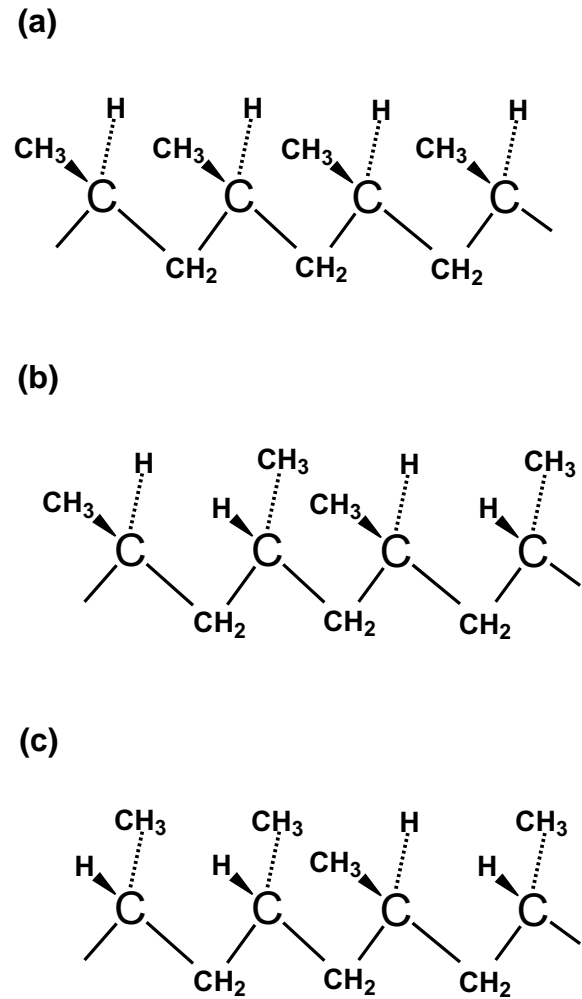


FIGURE 1 Stereochemical configuration of PP: (a) isotactic, (b) syndiotactic and (c) atactic

electrical and mechanical properties for HVDC cable insulation. In another investigation, PP/POE blends were considered [35]. It was shown that blending PP with POE resulted in desirable properties needed for HVDC cable insulation. POE content had little effect on the breakdown strength, where only 10% reduction in breakdown strength was reported when the content of POE was high (30 wt%). In addition, Hosier et al. [36] compared different blends of PE and PP to XLPE and found that PP-based blends outperformed XLPE in mechanical, thermal and electrical properties at higher temperatures. It was shown that PP-based blends offered 35.77% higher DC breakdown strength compared to XLPE.

It has been found in the study that copolymerization and blending can be helpful in achieving good mechanical properties of the overall PP, but the electrical properties reduce in most cases, which can be linked to the formation of space charge. As a consequence, the potential of PP-based materials for HVDC cable has not been fully realized. One way to enhance the breakdown strength of PP-based HVDC cable insulation can be through the use of nanotechnology.

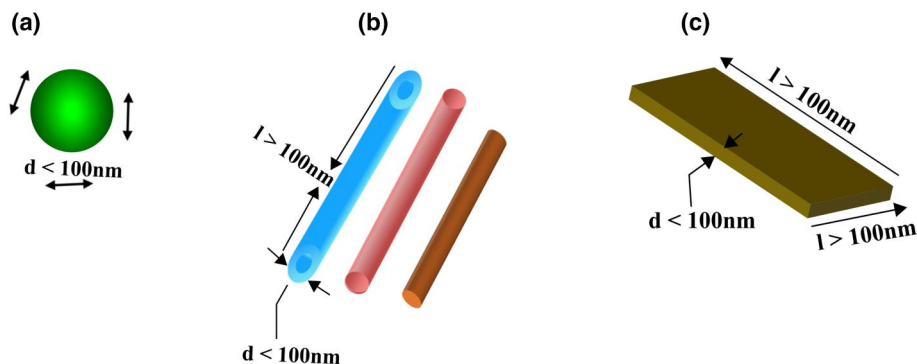


FIGURE 2 Classification of nanofillers based on dimensionality: (a) 0D-spherical nanofiller, (b) 1D-nanotubes, nanorods and nanowires and (c) 2D-nanosheets

4 | INFLUENCE OF NANOTECHNOLOGY ON POLYPROPYLENE-BASED NANOCOMPOSITES

The concept of nanometric dielectric in electrical insulation was brought about by Lewis in 1994, by exploring the role of nanometric size particle inclusion in polymers that would lead to the enhancement in the electrical insulation properties [10]. Later in 2001, Frechette et al. [37] proposed the term ‘nanodielectric’ as a multicomponent dielectric possessing nanostructures, the presence of which leads to changes in one or several of its dielectric properties. Nanocomposites differ from conventional composites in the following aspects [38]: (i) the filler size is in the range of nanometric scale (usually less than 100 nm) compared to microfillers, (ii) nanofillers are usually incorporated in a small loading (less than 10 wt%) compared to microfillers which are more than 50 wt% for microcomposites and (iii) nanofillers have very large specific surface area compared to microfillers. To date, different kinds of nanofillers have been added to PP-based polymers in order to enhance its properties.

4.1 | Classification of nanofillers used in nanodielectrics

The dimension and shape of a nanofiller are important. Nanofillers are classified on the basis of their dimensions which are not confined to the nanoscale range (<100 nm) (Figure 2) [39]: 0D, 1D and 2D.

4.1.1 | Zero-dimensional nanofillers

These fillers are also known as isotropic nanomaterials. 0D nanofillers are the class of nanomaterial in which all dimensions are nanoscale, that is, the width, length and thickness of these nanofillers are within the range of nanometric scale. These nanofillers are also called nanosphere or nanocrystal. 0D nanofillers are usually found in spherical or cubical shape. Examples of this type of nanofillers include titania (TiO_2),

silica (SiO_2), alumina (Al_2O_3), zinc oxide (ZnO), magnesia (MgO), zirconia (ZrO_2), aluminium nitride (AlN), boron nitride (BN) etc.

4.1.2 | One-dimensional nanofillers

Fillers in which at least two dimensions are in nanometric range while one dimension (length) is outside the nanometric scale are called 1D nanofillers. These nanofillers are called as anisotropic nanofillers. These nanofillers include nanowires, nanofibres and nanotubes. Carbon nanotube, hexagonal boron nitride tube and 1D metal oxide (e.g. TiO_2 , ZnO and Al_2O_3) are the most common 1D nanofillers used.

4.1.3 | Two-dimensional nanofillers

The nanofillers in which at least one dimension is in nanometric range are called 2D nanofillers. These nanofillers are called as anisotropic nanofillers. These fillers exhibit in-layer or sheet form with thickness in the range of few nanometres. 2D nanofillers include nanosheets and nanoplates. Nanoclay (layered silicate) and graphene nanoplatelet are a typical example of 2D nanofiller.

4.2 | Surface modification of nanofillers

The surface of nanofillers is commonly modified in order to achieve homogenous dispersion of nanofillers in polymer matrices, which can help in hindering the nanofillers from agglomeration and making the nanofillers surface more compatible with that of polymer matrices by overcoming the immiscibility between hydrophilic fillers and hydrophobic polymers. Surface modification of nanofillers can be achieved using different approaches [40]: (i) chemical surface modification and (ii) grafting synthetic polymer on the surface of nanofillers.

In chemical surface modification of nanofiller, the modifier is covalently bonded onto the surface of nanofillers, resulting in a strong interaction between modifier and nanofiller. Chemical surface modification of nanofiller can be achieved

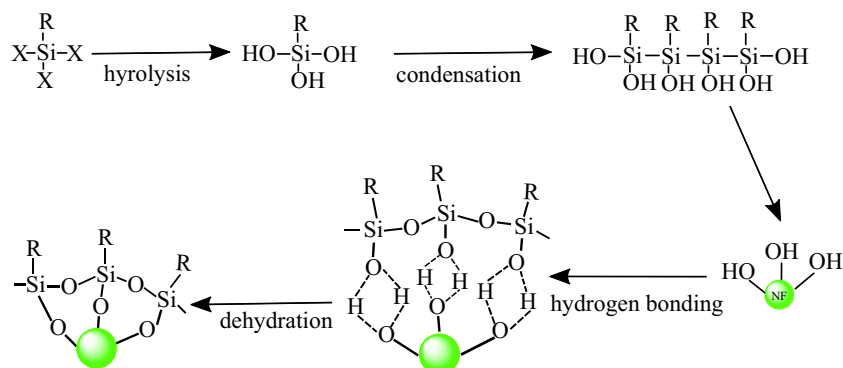


FIGURE 3 Surface modification mechanism of inorganic nanofiller using silanes

using coupling agents. Coupling agents like silane, titanate [41] and zirconate [42] have been applied to change the surface of hydrophilic fillers. Silane coupling agents are most widely used coupling agents for surface modification of inorganic nanofillers. The general structure of silane coupling agent can be represented as $\text{R}-(\text{CH}_2)_n-\text{Si}-\text{X}_3$, where R represents the organofunctional group (amino, methacryle, epoxy) which reacts with the polymer, $(\text{CH}_2)_n$ represents the linker length, which plays an important role in properties of the coupling system while X represents the hydrolysable group (e.g. methoxy, ethoxy) that reacts with the surface of the nanofiller. Surface modification of nanofillers commonly takes place in liquid media by adding nanofiller and silane coupling agent in a common solvent (e.g. ethanol, acetone, xylene, toluene). The surface of the metal nanofillers is usually hydrophilic with hydroxyl groups ($-\text{OH}$) on it which is mainly used as a reaction site. During the silane coupling modification, the $-\text{OH}$ group of nanofiller reacts with the hydrolysable group of the silane coupling agent. The silane modification usually involves four steps (hydrolysis, condensation, adsorption and chemical grafting) [43]. During hydrolysis process, the alkoxy groups attached to silicon atom of silane coupling agent are hydrolyzed, forming a reactive silanol groups ($\text{Si}-\text{OH}$). In the condensation step, the condensation of reactive silanol happens and this results in the formation of silanol oligomers. During adsorption, the reactive silanol oligomers are physically attached to the hydroxyl groups of nanofillers via hydrogen bonding on the surface of nanofillers. In the chemical grafting process, drying or curing is applied which results in the formation of a covalent bond between the metal nanofiller and the silicon atom of silane coupling agent. The R groups from the attached silanes are available to interact with different polymer chains via covalent linkage or physical interaction. The surface-modified nanofillers are then separated from the solvent using different methods such as filtration, decantation or centrifugation and then usually washed with the solvent several times to remove the excess silane coupling agents. An example of the process of silane modification of organic nanofillers is illustrated in Figure 3 [44]. The materials can be SiO_2 particles treated with trimethoxy-(propyl) silane. It was reported that the breakdown strength of PE/ SiO_2 nanocomposites increased upon incorporation of silane-modified SiO_2 as compared to unmodified SiO_2 [45].

Another technique to modify the surface of nanofiller is grafting polymer brushes on the surface of nanofillers. Generally, grafting of polymer brushes includes two strategies to build interfaces between polymer brushes and nanofillers. The first strategy involves ‘grafting to’ approach, in which the as-prepared polymer brushes directly react with the functional groups present on the surface of the nanofillers. The ‘grafting to’ approach is simpler but it is difficult to obtain high graft density due to the steric repulsion of already attached polymer brushes for incoming polymer brushes. The second strategy involves ‘grafting from’ approach, in which in situ polymerization of monomers occur from the initiating groups present on the surface of nanofillers. Through this approach, a much higher density of graft polymers can be obtained on the surface of nanofiller by initiating the graft polymerization from initiating groups placed on the nanofillers’ surfaces. For example, Zhang et al. [46] grafted poly (stearyl methacrylate) (PSMA) polymer on the surface of nano- SiO_2 using grafting from approach. It was reported that PSMA grafted SiO_2 nanofillers dispersed homogeneously in XLPE, which resulted in improved electrical properties of XLPE/ SiO_2 nanocomposites.

4.3 | Impact of interfacial area on the properties of nanocomposites

The addition of nanofillers to polymer matrices can give rise to superior electrical, thermal and mechanical properties that the polymers alone cannot achieve. It is believed that the interface region (in nanometric scale) found between the nanofiller and the polymer in polymer nanocomposites is responsible for such properties improvements, as the nanocomposite interfacial area possesses superior properties that belong neither to the base polymer nor the filler [47]. Therefore, several attempts have been made to better understand the nanocomposite interface properties and its physicochemical structure, subsequently leading to different interface models as illustrated in Figure 4. Although different interface models have different viewpoints with regard to the interfacial area, they all share a common concept: the interface layer closest to the nanofiller, which is commonly characterized by nanofiller surface modification, is key to controlling the behaviour of the interfacial area and affecting the properties of nanocomposites [48].

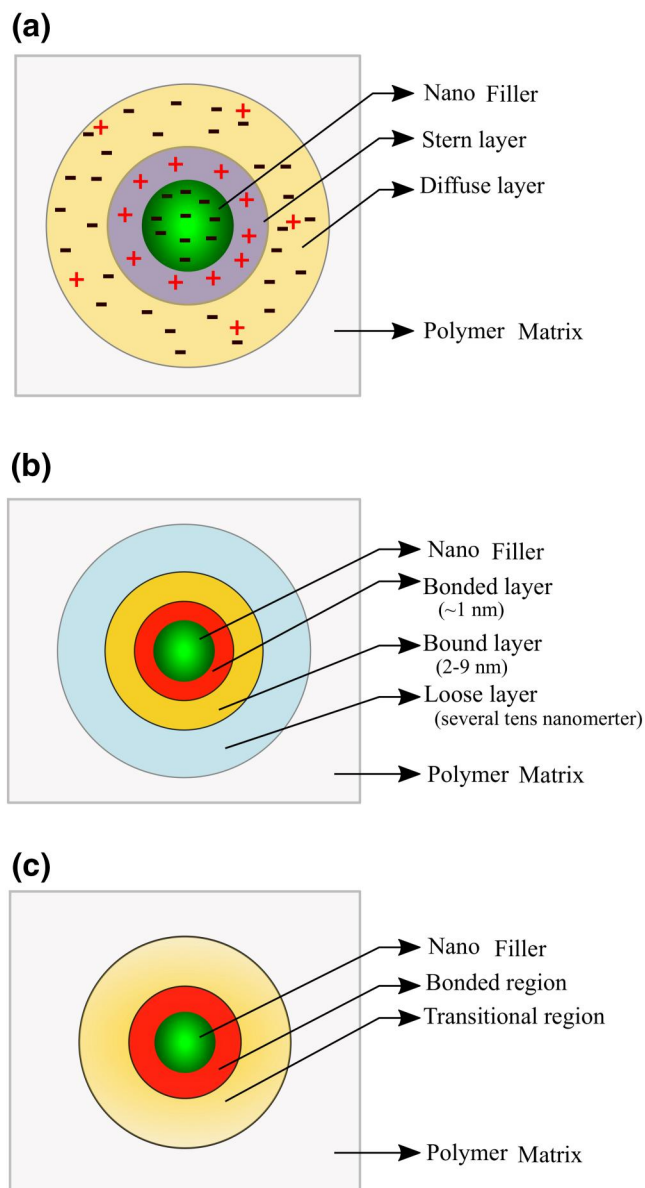


FIGURE 4 Schematic illustration comparing the interface between a nanofiller and a polymer: (a) electrical double layer model, (b) multicore model and (c) multiregion structure model. Reillustrated from [10, 51, 52]

4.3.1 | Electrical double layer model

Lewis emphasized the significance of the nanocomposite interfaces by proposing the ‘electrical double layer model’ [10, 49, 50]. According to Lewis, interfaces are the region with different electrochemical and electromechanical properties compared to the polymer matrix and the nanofiller. When nanofillers are incorporated in a polymer, a Stern and Gouy–Chapman diffused layers are formed at the interface between the nanofiller and the polymer (Figure 4(a)) [49]. The Stern layer is formed at the exterior of the nanofiller and is tightly bonded to the surface of nanofiller. Meanwhile, the second layer, the diffused layer, is formed due to the distribution of negative and positive ions. This layer determines the dielectric

properties of nanocomposite as it potentially works as an interaction zone. Lewis suggested that when the nanofiller content is low, the electrical double layer disrupts the movement of charge carrier thus resulting in enhanced breakdown strength. As the loading further increases the interfacial region may overlap, which leads to the formation of a conduction path within the nanocomposites, thus promoting the acceleration of free charge carriers and eventually results in a decrease of breakdown strength.

4.3.2 | Multicore model

In 2005, Tanaka proposed the multicore model [51]. The model determines the chemical and electrical features of the interface region formed between the spherical nanofiller and the polymer matrix (Figure 4(b)) [51]. According to Tanaka’s model, the interface region is defined as multilayers of several tens of nanometre and consists of three layers, namely, first layer (bonded layer), second layer (bound layer) and third layer (loose layer). Beside these layers, Tanaka also proposed the presence of the fourth layer (Gouy–Chapman diffused layer) that overlaps the above three layers when Coulombic interaction is superimposed. The thickness of this layer is estimated from several tens to 100 nm [53]. The first layer, having thickness of around 1 nm, is a transition layer which is tightly bonded to the nanofiller and polymer matrix. Usually, coupling agents such as silanes are used to couple the inorganic nanofiller (polar) and organic polymeric matrix (nonpolar). Primarily, the first layer is bonded by ionic, covalent, hydrogen and Van der Waals bonding. The second layer, having thickness in the range of 2–9 nm, represents an interfacial region consisting of a layer of polymer chains that are strongly bonded to the first layer. Deep traps exist in both the bonded and bounded layer, which help in the enhancement of breakdown strength of nanocomposites. The third layer, having thickness >10 nm, has different chain confirmation, chain mobility and even free volume or crystallinity from the polymer. This layer is loosely bonded with the second layer. With the help of multicore model, several dielectric properties, such as change in permittivity, space charge suppression, dielectric breakdown strength and partial discharge can be explained [54].

4.3.3 | Multiregion structure model

The multiregion structured model was proposed by Li et al. [52]. It describes the formation of a multiregional structure around the surface of modified and unmodified nanofillers in polymer nanocomposites. As reported in this model, the interface between the nanofiller and the polymer consists of the bonded region and the transitional region. The normal region represents the polymer matrix, as shown in Figure 4(c). The bonded region is rich in deep traps, and is formed when the nanofiller is coupled with the polymer matrix through covalent, ionic, hydrogen bonds and Van der

Waals bonding. It is assumed that this region has the strongest bonding strength due to the presence of unsaturated bonds, for example, hydrogen and organic groups (silane coupling), present at the surface of nanofiller. Meanwhile, the transitional region is situated next to the bonded region. It is postulated that the transitional region contained ordered polymer chains which strongly depend on the cohesive energy density (CED) of the polymer matrix. Higher CED of the polymer matrix results in better flexibility of the molecular chain and extended service life of the nanocomposites. Both the bonded region and transitional region have influence on the short-term and long-term breakdown properties of nanocomposites [52].

4.4 | Fabrication methods

Nanofillers should be homogeneously dispersed in polymers in order to achieve desirable properties. It is found that nanofiller agglomeration in polymer brings detrimental effect on the electrical properties of polymer nanocomposites. In general, when formulating polymer nanocomposites, the aim should be to achieve uniform particle distribution and minimal nanofiller agglomeration. For proper dispersion of nanofillers throughout the polymer matrix, a proper fabrication method should be adopted. Over the past decades, different approaches have been used for the fabrication of polymer nanocomposites. In general, nanofiller can be dispersed in polymer matrices using four different approaches: (i) interaction method, (ii) in situ polymerization, (iii) sol gel method and (iv) direct mixing of polymer and nanofillers (Figure 5) [55].

4.4.1 | Intercalation method

Intercalation process is usually followed for dispersing 2D nanofillers (platelet/sheet) into the polymer matrix. Intercalation method is a top down approach which usually involves the surface modification of plate/sheet nanofillers for the homogenous dispersion in polymer matrix. The intercalation of polymeric materials and exfoliation of 2D nanofillers can be achieved via chemical or mechanical routes. The chemical route involves in situ polymerization of monomers by organic initiators in the presence of layered nanofillers. During the process of polymerization, the monomers are polymerized between the interlayers of layer fillers forming either intercalated or exfoliated nanocomposites. On the other hand, the mechanical route involves the direct intercalation of polymers with 2D nanofillers. This can be achieved either by dispersing nanofillers in an appropriate solvent (such as toluene, chloroform or water) or melt intercalation via melt compounding in the absence of solvents. Among these approaches the melt intercalation is favourable because it does not involve the use of any solvents, which is preferable from an environmental point of view.

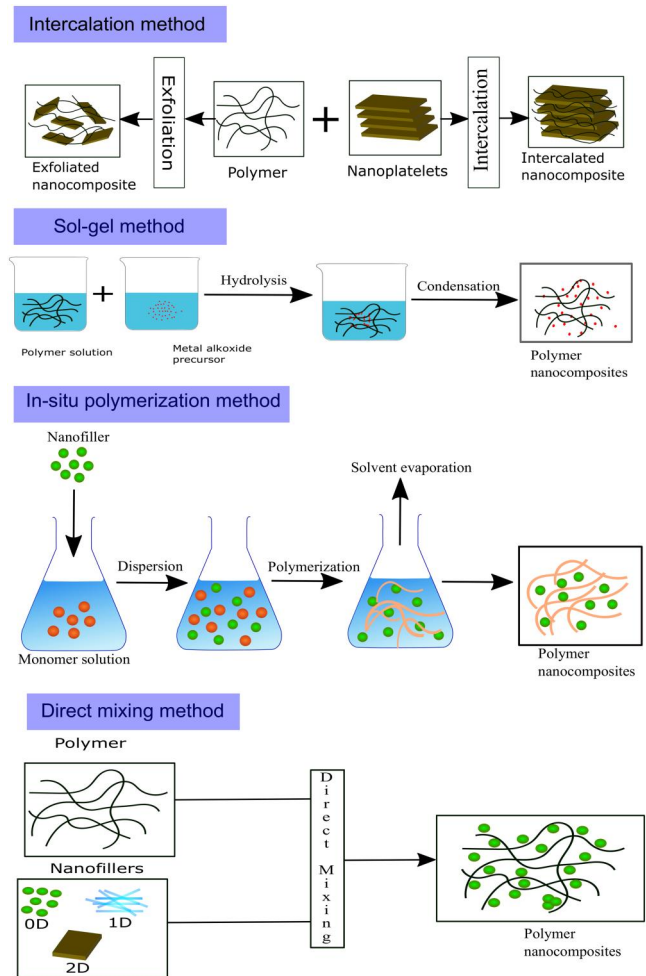


FIGURE 5 Schematic illustration of the main fabrication methods for polymer nanocomposites

4.4.2 | Sol gel method

The sol gel method is a conventional bottom up method. In this method, nanofillers are prepared in the presence of polymer. The nanofillers are formed by hydrolysis and condensation reactions of metal alkoxides. SiO_2/PP nanocomposites were prepared in [56]. PP was first dissolved in xylene, and tetraethyl orthosilicate is then added under suitable conditions.

4.4.3 | In situ polymerization

In this method, the nanofillers are properly dispersed in the monomer solution. During in situ polymerization, the monomers polymerized via standard polymerization methods in the presence of nanofillers. Käselau et al. [57] reported synthesis of a well dispersed magnesium hydroxide $(\text{Mg}(\text{OH})_2)$, SiO_2 and $\text{BaTiO}_3/\text{iPP}$ nanocomposites through in situ polymerization method. In their method the surface-modified nanofillers were dispersed in propylene and the polymerization was achieved using a suitable initiator.

4.4.4 | Direct mixing

The direct mixing approach is a typical top down approach based on the breakdown of nanofiller aggregates through the application of high shear force during mixing process. In this method the nanofillers are directly mixed with polymer matrix. Polymer nanocomposites can be prepared through this method by two approaches, that is, solvent mixing and melt compounding methods.

In the solvent mixing method, nanofillers are dispersed in polymer in the presence of solvent. The solvent mixing method usually consists of three steps, namely (a) dispersing the nanofiller in a suitable solvent, (b) mixing the nanofiller suspension with the polymer in the presence of the same solvent or cosolvent and (c) removing the unwanted solvent from the composite solution, either by evaporation or filtration. At the large scale, the major drawback of this method is that it is slow and requires a large amount of solvents and energy; this is an environmental unfriendly approach.

In the melt compounding method, nanofillers are directly mixed with polymers in the absence of solvents. Typically, the melt compounding process involves the heating of polymer above its melting temperature in the mixing equipment (e.g. a twin-screw extruder), followed by the addition of nanofillers under the application of high shearing force to obtain homogeneously dispersed nanocomposites. Due to recent improvements in mixing devices and surface treatment techniques, the melt blending method is shown to be the most technologically relevant method as it is a simple technique and does not require any solvent, and can be carried out on a large scale. The melt blend method is therefore widely adopted for the preparation of polymer nanocomposites in recent years both by industry and academia.

5 | RECENT DEVELOPMENT IN POLYPROPYLENE-BASED NANOCOMPOSITES

The incorporation of nanosize fillers into polymers can lead to the creation of enormous interfacial region which could result in an improvement in electrical properties of polymer nanocomposites that cannot be achieved through unfilled polymers and microcomposites [49]. Nevertheless, the improvement in electrical properties of polymer nanocomposites largely depends on several parameters such as nanofiller loading, nanofiller type, nanofiller dispersion and nanofiller morphology in polymer matrices. For PP nanocomposites, good electrical properties (high breakdown strength, suppressed space charge, lower DC conductivity), thermal properties (high thermal conductivity and thermal stability) and good mechanical properties are necessary. Till date, different kinds of nanofillers with different dimensions are added into PP matrices for enhancing the breakdown strength of PP-based nanocomposites. In the following sections, the effect of various parameters such as nanofiller loading, nanofiller type, nanofiller surface chemistry and nanofiller

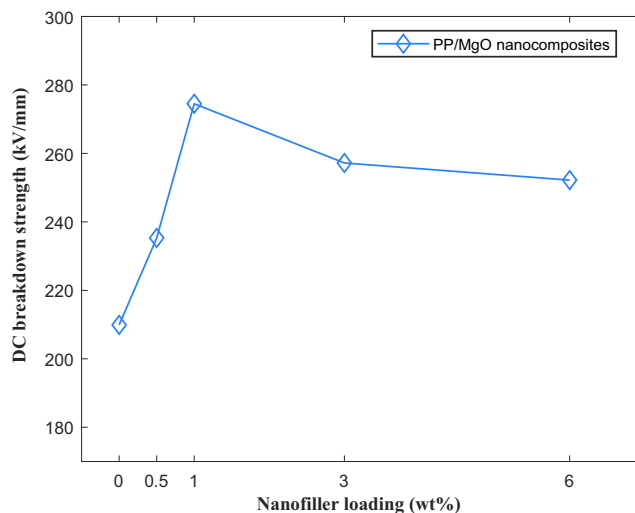


FIGURE 6 Direct current breakdown strength of polypropylene nanocomposites with different nanodoping contents. Compiled from [58]

morphology on the dielectric properties of PP-based nanocomposites is summarized.

5.1 | Effect of nanofiller loading

Usually, the dielectric properties of PP improved upon the addition of a small amount (0.5–3 wt%) of nanofillers. The effect of nanofiller content on the properties of PP-based nanocomposites has been reported in many studies. Cao et al. [58] reported the effect of varying loads of nano-MgO on the dielectric properties of PP/MgO nanocomposites. As illustrated in Figure 6, the DC breakdown strength increased when the nanofiller loading was increased until an optimum filler loading. When the filler loading was further increased beyond the optimum amount, the DC breakdown strength started declining. The decline in DC breakdown was attributed to the agglomeration of nanofillers at higher loading. Nevertheless, the DC breakdown strength of PP/MgO nanocomposites remained higher than that of unfilled PP for all nanocomposite systems [58]. Similarly, for low MgO loading the accumulated heterocharges near electrodes were suppressed as compared to neat PP. However, at loading of 6 wt%, due to higher agglomeration of MgO, PP/MgO nanocomposites have more space charge accumulation compared to neat PP. Similar results were demonstrated for PP/Al₂O₃ nanocomposites in [59], where the DC breakdown strength increased upon the addition of low loading of Al₂O₃ and reached at a maximum value at 3 parts per hundred of rubber (phr) with effective space charge suppression. However, the DC breakdown strength started to decrease as the loading of Al₂O₃ reached 5 phr and more space charge was accumulated.

It is worth nothing that, the electrical properties are dependent on the interparticle distance between the nanofillers in polymer matrix. When the filler content is increased beyond an optimum level, the mean particle distance between the fillers

will decrease, which can result in an overlap of the interface, thus resulting in the formation of tunnel to breakdown more easily. For instance, Zha et al. [60] showed that 0.5 phr MgO nanofiller enhanced the DC breakdown strength of PP/styrene-ethylene-butylene-styrene (SEBS)/MgO nanocomposites as compared to unfilled PP/SEBS blend. The authors attributed this improvement to the introduction of deep traps that were formed in the polymer/filler interface, which successfully captured the space charge and reduced the free charge carrier mobility. However, a further increase in MgO content (1–1.5 phr) led to the deterioration in DC breakdown strength and increased space charge accumulation. It was proposed that at higher loading of MgO, the trap density became too high, which resulted in ease of charge carrier movement between adjacent traps. Xie et al. [61] investigated the effect of higher filler loading on the DC breakdown strength and conductivity. At 1 wt% PP/Al₂O₃ nanocomposites, an increase of 36.7% DC breakdown strength was reported with reduced conductivity compared to pure PP. When the Al₂O₃ content increased to 7, 12 and 20 wt%, the DC breakdown strength decreased by 9.2%, 21.1% and 28.6%, respectively with high DC conductivity. The authors linked the reduction in breakdown strength to the overlapping interfaces at higher loading which facilitate carrier migration between neighbouring particles. In another study, Zha et al. [62] investigated the electrical and mechanical properties of PP/SEBS nanocomposites containing ZnO at different loadings (0.2, 0.5, 1 and 2 wt%). It was found that at 0.5 wt% of ZnO, the performance of PP/SEBS nanocomposites was improved. The tensile strength and elongation at break at 0.5 wt% increased to 18% and 24.7%, respectively compared to PP/SEBS blend. Similarly, at 0.5 wt %, an improved space charge suppression and DC breakdown strength were achieved with a lowered DC conductivity.

5.2 | Effect of nanofiller types

Every nanofiller possesses different physical properties from other nanofillers like electrical insulation, permittivity and thermal conductivity, which can influence the dielectric properties of polymer nanocomposites. For example, Zhou et al. [63] performed a comparative study on the effect of types and loadings of nanofillers on the electrical properties of PP composites. Four different inorganic nanofillers, namely, MgO, TiO₂, ZnO and Al₂O₃, having an average diameter of 50 nm were used. It was reported that inclusion of the nanofillers resulted in improved electrical properties of PP nanocomposites. The DC volume resistivity was reported to increase for the nanofillers with Al₂O₃ showing increasing resistivity with increasing content, the opposite was observed for TiO₂ at higher loading. MgO and TiO₂ showed the best performance in effectively suppressing the space charge and improving breakdown strength. The highest breakdown strength was reported with 1 and 3 phr TiO₂ and MgO (Figure 7), respectively. In another study, different loadings of Al₂O₃, TiO₂ and organoclay were incorporated in EPDM/PP blend and its effect on the DC breakdown strength was compared [64]. At 2 vol% loading, all the filler caused increase in

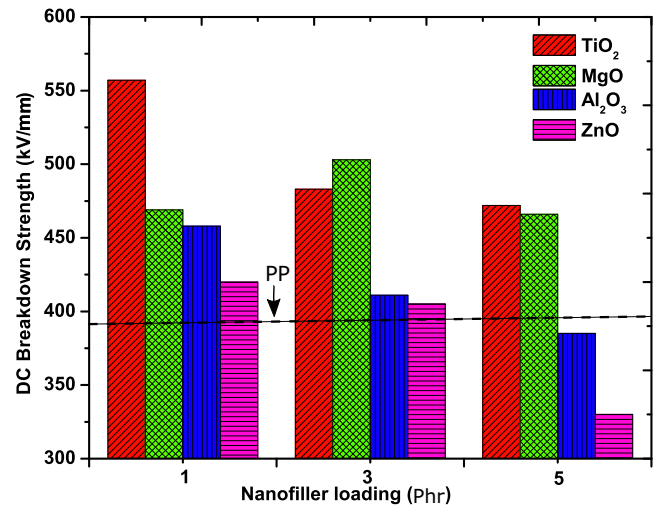


FIGURE 7 Direct current breakdown of PP nanocomposites with different nanofillers at different contents. Compiled from [63]

the breakdown strength while further increasing the loading cause decrease in the breakdown strength. Among the three nanofillers, organoclay gave the highest DC breakdown strength.

Improving the thermal conductivity of polymer is an important aspect in HVDC cable insulation. The improved thermal conductivity will help in achieving a uniform temperature gradient between the internal and external insulation of HVDC cable [65]. PP with low intrinsic thermal conductivity ($0.14 \text{ Wm}^{-1}\text{K}^{-1}$) makes it a poor thermal conductive material. Nanofillers with high thermal conductivity and electrical insulating characteristics are usually added to improve the thermal conductivity of PP-based materials. Among different nanofillers, AlN and BN are typical examples of thermally conductive and electrically insulating nanofillers which are used to increase the thermal conductivity of PP [14, 66]. For example, Zhou et al. [12] added AlN nanofiller into PP at different loadings. It was found that the inclusion of AlN nanofiller effectively improved the thermal conductivity and thermal stability of PP nanocomposites. Moreover, it was shown that the presence of AlN in PP led to a reduction in breakdown strength and an increase in DC conductivity. Although the electrical properties of PP/AlN nanocomposites declined compared to pure PP, PP/AlN nanocomposites demonstrated better electrical properties than XLPE and low density polyethylene insulation materials [12].

5.3 | Effect of nanofiller surface chemistry

Due to the hydrophobic nature of inorganic nanofillers, nanofillers tend to agglomerate in nonpolar hydrophobic PP matrix. As a result, homogeneous dispersion of nanofillers is hard to realize. Surface modification of nanofillers generally helps in achieving homogeneous dispersion of nanofillers in PP-based matrix, which can result in enhanced dielectric properties [67–70]. Diao et al. [71] studied the mechanical and electrical properties of iPP/POE-SiO₂ nanocomposites (with

and without surface modification). Three different surface modifiers, namely, polydimethylsiloxane (PDMS), octylmethylsiloxane and dimethyldichlorosilane, were used to modify the surface of SiO₂. It was shown that surface modification of SiO₂ resulted in enhanced properties of the nanocomposites compared to iPP/POE blend and unmodified nanocomposites. Among the differently modified SiO₂, PDMS-modified SiO₂ resulted in improved space charge suppression and higher breakdown strength compared to other systems. Moreover, the tensile strength was found unaffected by the incorporation of unmodified and modified SiO₂. However, the addition of unmodified SiO₂ resulted in significantly decreased elongation at break property of the iPP/POE nanocomposites. A significant enhancement in elongation at break was reported for iPP/POE containing PDMS-modified SiO₂. The study concluded that both the electrical and mechanical properties of PP nanocomposites largely depended on the surface chemistry of nanofillers.

The chain length of silane modifiers can also have an influence on the electrical properties of polymer matrix [72, 73]. The effect of silane chain length on the dielectric breakdown strength of PP nanocomposites was studied by Hu et al. [74]. They selected trimethoxy silane with varying length of alkyl groups: methoxy (C1), propyl (C3), octyl (C8) and octadecyl (C18) to modify the surface of MgO nanofiller. The silane modification helps the MgO nanofiller to disperse well in the PP matrix, which could be attributed for better compatibility between modified MgO and PP. It was observed that the silane-modified MgO nanofillers were well dispersed in PP with less aggregation of MgO. All the nanocomposites showed effective space charge suppression and enhanced dielectric breakdown strength compared to pure PP. The highest dielectric breakdown strength (27.4% higher than pure PP) was reported for 3 wt% MgO modified with C8 silane. The results suggested that the C8 silane-treated MgO can induce more deep charge traps that can effectively capture the charge carriers and can decrease their energy and mobility which lead to the improvement of dielectric breakdown strength [74]. The amount of silane used for the surface modification of nanofiller can also influence the dielectric breakdown strength of polymer nanocomposites [75]. Insufficient amount of silane coupling agent may lead to less coverage of nanofiller which can result in poor dispersion of nanofiller in polymer matrix. On the contrary, excessive amount of silane can undergo a self-condensation reaction, which could degrade the electrical properties of nanocomposite systems. Wang et al. [76] selected different amounts of trimethoxy (octyl) silanes based on an estimation method for the modification of AlN nanofiller. Their results suggested that the amount of silane has little effect on the dielectric breakdown strength of PP/AlN nanocomposites. Krentz et al. [77] used polymer grafting technique for modifying the surface of nano-SiO₂. They grafted anthracene and PSMA on the surface of SiO₂. Anthracene, being the smaller molecule, acts as a voltage stabilizer that helps in creating deep traps for charge carriers while the long PSMA brushes help in dispersing the anthracene attached SiO₂ well in PP matrix. It

was shown that the combination of 2 wt% PSMA anthracene-modified SiO₂ increased both the AC and DC breakdown strength of PP/SiO₂ nanocomposites. The increase in breakdown strength was ascribed to the better dispersion brought by PSMA brushes and the charge trapping characteristics of anthracene.

Since heat transfer in polymer is done through phonon, weak coupling of interface between polymer matrix and nanofillers can offer high interfacial resistance resulting in a strong phonon scattering which can reduce thermal conductivity of polymer nanocomposites [78]. Surface modification of nanofillers helps in improving the compatibility between nanofillers and polymer matrices, thus decreasing the interfacial resistance between the polymer matrices and the nanofillers. Wang et al. [13] investigated the effect of filler surface chemistry on the thermal conductivity of PP/AlN nanocomposites. Three different silanes (3-trimethoxysilyl-propyl methacrylate, trimethoxy(octyl)silane and triethoxy(octyl)silane) having varying organofunctional and organosoluble groups were used to modify the surface of AlN. The silanes treatment improved the compatibility of filler and PP matrix which results in reduced phonon scattering. The authors reported 15% improvement in thermal conductivity of the nanocomposite material when 10 wt% 3-trimethoxysilyl-propyl methacrylate silane-treated nano-AlN was added into PP. The improvement was linked to the presence of organic layer on the AlN nanofiller which weakened the phonon scattering and decreased the thermal interfacial resistance.

5.4 | Effect of nanofiller morphology

The dielectric properties of nanocomposites also depend on the geometry of nanofillers. Beside 0D nanofillers, 1D and 2D nanofillers are also incorporated into PP for tailoring its dielectric properties [79–87]. Since the anisotropy of 1D and 2D nanofillers is different from 0D, they result in different interface adhesions. Recently, graphene-based nanofillers have been used in many research studies for enhancing the electrical properties of polymer HVDC nanocomposites. The high specific surface area of graphene nanoplatelet (2000 m²/g) helps in the creation of enormous interfacial area between polymer and nanofiller. Graphene can act as a voltage stabilizer that can help in dissipating the high energy of hot electron originated from electrodes that may damage the polymer chains. Du et al. [79] incorporated a very low content of 2D graphene nanoplate (diameter: 0.2~10 μm, thickness: ~1 nm) into PP/ultra low density polyethylene (ULDPE). ULDPE was blended with PP at different ratios to obtain good mechanical properties tailored for HVDC cable insulation. The best combination obtained was 15 wt% ULDPE. Upon incorporation of a very small content of graphene nanoplatelets, much lower DC conductivity, enhanced breakdown strength and effective space charge suppression compared to neat PP and PP/ULDPE blends was achieved. The authors ascribed improvements in electrical properties due to the presence of graphene nanoplatelets that led to the

creation of massive deep traps in the interfacial region which helps in restricting the charge carrier moment, and providing a higher potential barrier for charge carriers to overcome. Similarly, Zhang et al. [83] studied the effect of 2D boron nitride nanosheet (BNNS) on the electrical and thermal properties of PP/SEBS nanocomposites. It was found that the inclusion of 3 phr BNNS into PP/SEBS resulted in effective space charge suppression and the breakdown strength was 11% higher than PP/SEBS blend. Moreover, BNNS inclusion enhanced the thermal stability of PP/SEBS nanocomposites. The thermal conductivity of PP/SEBS nanocomposites increased 3 times at 3 phr BNNS compared to PP/SEBS blend. The improved thermal properties were due to the high aspect ratio of BNNS which formed perfect continuous heat conductive pathways in polymer matrix. Nanoclay is also used to enhance the breakdown strength and suppress the space charge of PP. For instance, Eesaee et al. [86] investigated the AC breakdown strength of PP/SEBS blend nanocomposites containing 2D organomodified clay. It was reported that, by the addition of 5 wt% organoclay, an improvement of 25% was achieved in breakdown strength with suppressed space charge. The authors linked the improvement to the plate-like shape of nanoclay structure that formed a tortuous path for the charge carrier to travel across the nanocomposites.

It should be noted that the orientation of nanofillers also plays an important role in controlling the electrical properties of polymer nanocomposites [88, 89]. For example, the effect of 2D organo-montmorillonite (o-MMT) on the breakdown strength of polymer nanocomposites was studied [89]. It was shown that 6 wt% high oriented o-MMT resulted in an increased breakdown and lowered DC conductivity compared to neat polymer and randomly dispersed o-MMT. The improved performance was attributed to the ability of oriented 2D o-MMT that created a prolonged tortuous path for free electrons to mobilize.

Metal oxide nanofillers can be found in different shapes and sizes (0D, 1D and 2D). Zha et al. [90] studied the effect of ZnO nanofiller's shape on the DC breakdown strength of PP. 1 wt% of three different ZnO shapes, namely ZnO nanoparticle, ZnO nanowire and ZnO nanowhisker, were incorporated into PP. Comparing with neat PP, the ZnO nanoparticle and ZnO nanowire increased the breakdown strength of PP nanocomposites. The breakdown strength decreased in case of PP nanocomposites added with ZnO nanowhisker. The highest improvement was brought about by ZnO nanowires, which were attributed to the structure of ZnO nanowire capable of introducing more charge carrier traps. In another study, PP was added with two types of nano-TiO₂ (0D nanosphere and 1D nanorod) [91]. It was experimentally shown that the breakdown strength of PP filled with spherical TiO₂ was much higher than the PP filled with TiO₂ nanorods. The authors attributed the decrease in breakdown strength of PP filled with TiO₂ nanorod to the enhanced local electric field around TiO₂ rods, which can facilitate charge injections.

5.5 | Effect of temperature

Temperature also has significant effects on the performance of PP-based HVDC insulation. The operating temperature of insulation materials can change with varying loads, and this can affect the performance of cable insulation materials. It has been shown that the electrical properties (DC conductivity, space charge and breakdown strength) are affected at high temperature. Therefore, enhancing the electrical properties of polymers under higher temperature is much necessary. It has been found that the addition of nanofillers in PP-based materials resulted in an improved performance at high temperatures [16, 22, 81, 92]. For example, the effect of temperature on the electrical properties of PP and PP/MgO at different loadings (1, 3 and 5 phr) was studied in [22]. The results demonstrated that pure PP showed much dependence on temperature. With increased temperature from 30°C to 90°C, the DC conduction current increased while the DC volume resistivity decreased. Moreover, at higher temperature above 70°C more heterocharges were accumulated in pure PP material. Similarly, the breakdown strength decreased with increasing temperature. Nevertheless, the addition of nano-MgO led to better temperature dependent properties of PP nanocomposites compared to pure PP. It was shown that at 3 phr MgO, the DC volume resistivity at 90°C was found to be $10^{14} \Omega\text{m}$, which was about two orders of magnitude higher than the traditional XLPE. Additionally, space charge was effectively suppressed at all temperature ranges and an increase of 30% in breakdown strength was reported compared to pure PP in the temperature range from 30°C to 90°C. In another study, Du et al. [81] demonstrated that the addition of a small content of 2D graphene nanoplatelets (0.01%) in PP/ULDPE nanocomposites led to enhanced performance of space charge, DC conductivity and breakdown strength at a temperature range from 30°C to 90°C. These results indicate that nanofiller addition in polymer matrices has the potential to effectively suppress the space charge, increase breakdown strength and reduce DC conductivity both at room temperature and at higher temperatures compared to neat polymer.

6 | CONCLUSIONS AND FUTURE PERSPECTIVES

The recent developments in the field of HVDC insulation demand for a high performance ecofriendly insulation materials. PP, due to its superior electrical and thermal properties, has gained great attention in the field of HVDC cable insulation and is expected to be a good alternative in replacing conventional XLPE in HVDC cable insulation. However, research on PP-based HVDC cable insulation materials is at initial stages and there are still some limitations of PP-based materials such as low thermal conductivity, low-temperature impact performance, space charge accumulation and ageing under high temperature and DC stresses. Therefore, the development of PP-based HVDC insulation with optimized

electrical, thermal and mechanical properties is imperative in discovering a potentially recyclable cable insulation material. Nanodielectrics are considered as a promising approach for enhancing the dielectric properties of polymers. The addition of nanofillers in polymer results in a huge interfacial area in polymer nanocomposites that play a dominant role for nanocomposites properties.

In this review, the limitation of conventional XLPE insulation is presented. The structure, advantages and properties of PP-based insulation materials are discussed. Moreover, discussion on different types of nanofiller used in polymer nanocomposites is presented. Surface modification of nanofillers and preparation methods of nanocomposites are reviewed in detail. Several nanodielectric models discussing the structure and properties of interface between polymer and nanofiller are presented and compared. It is observed from the review of several studies that various parameters including nanofiller loading, types, surface chemistry, and morphology and temperature have substantial influence on the dielectric properties PP nanocomposites. The addition of low loading of nanofiller can bring improvements in the dielectric properties of PP nanocomposites. For enhancing the electrical properties, the content of nanofiller should not exceed the optimum level in order to avoid overlapping of particles. Various types of inorganic nanofillers with different physical properties and morphologies have been incorporated into PP which brings about different properties changes in PP nanocomposites.

For HVDC insulation, the selection of right choice of nanofillers is essential. Nanofillers which result in improved electrical properties (effective space charge suppression, high breakdown strength and low DC conductivity), improved thermal properties and at the meantime maintain the good mechanical properties should be the choice for PP nanocomposites. Although nanofiller inclusion brings improvement in the dielectric properties of nanocomposites, more investigations are required on the role of the nanofiller inclusion especially in correlating the different properties of nanofiller with the dielectric properties of nanocomposites. To enhance the dielectric properties of PP nanocomposites, the incorporation of the right nanofiller and optimal surface modification conditions for nanofillers have to be appropriately determined. Specifically, controlled synthesis of nanofillers in a laboratory scale is an attractive approach to produce a set of similar nanofillers but with differently tailored characteristics. This will allow a better structure–property relationship analysis of the synthesized PP-based nanocomposites. Of note, it can be a challenge to achieve homogenous dispersion of nanofiller in polymer matrices especially at the industrial scale. Therefore, the development of advanced material mixing tools and chemical formulation strategies in transitioning research achievements at the laboratory scale to the industrial scale will be crucial in determining successful implementation of PP nanocomposites as future HVDC cable insulation.

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